

OFFICE OF NAVAL RESEARCH

Contract N00014-90-J-1193

TECHNICAL REPORT No. 3

Nonlinear Optical Processes in One-Dimensional Polymers

by

Xiao-shen Li, D. L. Lin and Thomas F. George

Prepared for Publication

in

Laser Surface Microprocessing

Edited by V. I. Konov, B. S. Luk'yanchuk and I. W. Boyd

Proceedings of the Society of Photo-Optical Instrumentation Engineers

Departments of Chemistry and Physics
State University of New York at Buffalo
Buffalo, New York 14260

December 1989

Reproduction in whole or in part is permitted for any purpose of the
United States Government.

This document has been approved for public release and sale;
its distribution is unlimited.

DTIC
SELECTED
DEC 26 1989
S D

89 12 26 075

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1a. REPORT SECURITY CLASSIFICATION Unclassified		1b. RESTRICTIVE MARKINGS	
2a. SECURITY CLASSIFICATION AUTHORITY		3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release; distribution unlimited	
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE			
4. PERFORMING ORGANIZATION REPORT NUMBER(S) UBUFFALO/DC/89/TR-3		5. MONITORING ORGANIZATION REPORT NUMBER(S)	
6a. NAME OF PERFORMING ORGANIZATION Depts. Chemistry & Physics State University of New York	6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION	
6c. ADDRESS (City, State, and ZIP Code) Fronczak Hall, Amherst Campus Buffalo, New York 14260	7b. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217		
8a. NAME OF FUNDING/SPONSORING ORGANIZATION Office of Naval Research	8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER Contract N00014-90-J-1193	
8c. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217	10. SOURCE OF FUNDING NUMBERS PROGRAM ELEMENT NO. PROJECT NO. TASK NO. WORK UNIT ACCESSION NO.		
11. TITLE (Include Security Classification) Nonlinear Optical Processes in One-Dimensional Polymers			
12. PERSONAL AUTHOR(S) Xiao-shen Li, D. L. Lin and Thomas F. George			
13a. TYPE OF REPORT	13b. TIME COVERED FROM _____ TO _____	14. DATE OF REPORT (Year, Month, Day) December 1989	15. PAGE COUNT 13
16. SUPPLEMENTARY NOTATION Prepared for publication in <u>Laser Surface Microprocessing</u> , Edited by V. I. Konov, B. S. Luk'yanchuk and I. W. Boyd, Proceedings of the <u>Society of Photo-Optical Instrumentation Engineers</u>			
17. COSATI CODES FIELD GROUP SUB-GROUP		18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number) ONE-DIMENSIONAL POLYMERS LASER-INDUCED SUSCEPTIBILITY NONLINEAR OPTICS OPTICAL STARK BLUE SHIFT POLYDIACETYLENE-TOLUENE SULFONATE THEORY	
19. ABSTRACT (Continue on reverse if necessary and identify by block number) New phenomena are observed theoretically in nonlinear optical responses of polydiacetylene-toluene sulfonate (PTS) irradiated by a strong laser beam. Within a two-level model of PTS, the transient behavior of the induced susceptibility is investigated. Optical nutation is found, and the optical Stark blue shift and bleaching are also found in qualitative agreement with experiments. In the steady state, a new type of optical tristability mediated by phonons is found. A possible mechanism responsible for this tristability is discussed.			
20. DISTRIBUTION/AVAILABILITY OF ABSTRACT <input checked="" type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT <input type="checkbox"/> DTIC USERS		21. ABSTRACT SECURITY CLASSIFICATION Unclassified	
22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. David L. Nelson		22b. TELEPHONE (Include Area Code) (202) 696-4410	22c. OFFICE SYMBOL

Nonlinear optical processes in one-dimensional polymers

Xiao-shen Li, D. L. Lin and Thomas F. George

Department of Physics & Astronomy
State University of New York at Buffalo
Buffalo, New York 14260

ABSTRACT

New phenomena are observed theoretically in nonlinear optical responses of polydiacetylene-toluene sulfonate (PTS) irradiated by a strong laser beam. Within a two-level model of PTS, the transient behavior of the induced susceptibility is investigated. Optical nutation is found, and the optical Stark blue shift and bleaching are also found in qualitative agreement with experiments. In the steady state, a new type of optical tristability mediated by phonons is found. A possible mechanism responsible for this tristability is discussed.

1. INTRODUCTION

Nonlinear optical processes have been employed to realize light frequency conversion, light modulation and switching, optical logic and memory storage in optical computing, signal processing and image analysis.¹ Because of such immense potential applications, it has been one of the most active areas of research for both basic understanding of the mechanism and technological development of electro-optical or all-optical devices for communication and computer systems.

Organic polymers are known to exhibit giant nonlinear optical susceptibilities with fast response time.² In particular, the third-order susceptibility $\chi^{(3)}$ of conjugated polymers has attracted much attention. It is expected to play an important role in phase conjugation, optical bistability and all-optical modulation.

The large optical nonlinearities in conjugated polymers, first predicted over ten years ago,³ have been observed in numerous systems in recent years. The largest $\chi^{(3)}$ is recorded for trans-polyacetylene⁴ (CH_x) for which a number of theoretical investigations have been carried out very recently. On the basis of a one-dimensional model,^{5,6} $\chi^{(3)}$ has been calculated for (CH_x) by means of the Keldysh Green's function,⁷ and for pristine (CH_x) with neutral solitons.⁸ Furthermore, the chain-length effects on $\chi^{(3)}$ for (CH_x) have also been studied.

Meanwhile, nonlinear optical responses in polydiacetylene (PDA) have attracted much attention. The nonlinear refraction index n_2 of PTS films¹⁰ and PDA-4BCMU¹¹ have been measured, and phonon-mediated excitonic processes in PTS have been observed in transient nonlinear optical response^{12,13} and in an optical Stark effect experiment.¹⁴

We report, in this paper, theoretical studies of the transient nonlinear optical response of virtual excitons in PTS and a new type of optical tristability of PTS in an optical cavity.

A-1

2. TRANSIENT OPTICAL RESPONSE OF PTS

The investigation of coherent nonlinear optical processes caused by semiconductor excitations in the transparent region well below the absorption edge^{15,18} has been extended to polymers.^{12,14} It is known that virtual excitons are created when the frequency of an incident laser beam is tuned below the exciton resonance. Such excitonic effects can be explored more naturally by probing the susceptibility¹⁸ and looking for transient changes that persist as long as the excitation. Provided that the sample is strongly excited well below the conduction band edge, the real part of the susceptibility can be examined by measuring the index of refraction, and the imaginary part is explored by the absorption spectrum.

As PDA (and, in turn, PTS) possesses fairly large $\chi^{(3)}$ and small transmission loss α , which can be reduced to as low as¹⁹ $\alpha < 1 \text{ db/cm}$, PDA can have a rather large ratio $\chi^{(3)}/\alpha$.²⁰ In addition, PDA is relatively easy to use in the construction of waveguides. In this section we take PTS as an example and study the transient behavior of its induced optical susceptibility due to irradiation by a strong pumping laser and a weak probing laser. The two lasers may have pulse duration of femtoseconds to picoseconds. For the PTS system we adopt a two-level model to describe its ground state and exciton state. The lifetime of the exciton is also introduced phenomenologically.

2.1 Two-level model

The two states of PTS are denoted by $|+\rangle$ and $|-\rangle$. We assume a dipole transition and define the dipole operators

$$S^- = |-\rangle\langle+| , \quad (1a)$$

$$S^+ = |+\rangle\langle-| . \quad (1b)$$

The inversion operator is given by

$$S^z = \frac{1}{2} (|+\rangle\langle+| - |-\rangle\langle-|) . \quad (2)$$

For simplicity, the effect of phonon mediation²² is ignored here. (Its inclusion will be discussed and published elsewhere.) Thus we consider a single virtual exciton and write the Hamiltonian in the rotating-wave approximation as

$$H = H_0 + H' \quad (3a)$$

$$H_0 = \hbar\omega_x S^z - \frac{1}{2} (\mu_o E_p e^{i\omega_p t} S^- + h.c.) \quad (3b)$$

$$H' = -\frac{1}{2} (\mu_o E_t e^{i\omega_t t} S^+ + h.c.) , \quad (3c)$$

where ω_x is the exciton frequency, μ_o is the dipole matrix element, and E_p and E_t are the amplitude and frequency of the pump and test fields, respectively. Here we have neglected the momentum dependence of virtual excitons.^{13,23} This Hamiltonian uses dipole or projection operators to describe the dipole transition between the ground state and the excitonic state of the material by external fields.

It is different from that of Ref. 13, where creation and annihilation operators were used for virtual excitons.

We assume further that initially the system is in its ground state with no virtual exciton present. Thus we have $|\psi(0)\rangle = |-\rangle$ at $t = 0$. For any later time $t > 0$, the state can be represented by

$$|\psi(t)\rangle = C_+(t)|+\rangle + C_-(t)|-\rangle . \quad (4)$$

Plugging (4) into the Schrödinger equation with the Hamiltonian (3), we find the equations satisfied by the amplitudes C_{\pm} in the rotating frame as

$$\dot{C}_+(t) = i\Omega_p^* e^{-i\Delta_p t} C_- + i\Omega_t^* e^{-i\Delta_t t} C_- - \gamma C_+ \quad (5a)$$

$$\dot{C}_-(t) = i\Omega_p^* e^{i\Delta_p t} C_+ + i\Omega_t^* e^{i\Delta_t t} C_+ , \quad (5b)$$

where we have defined $\Omega_p^* = \mu_p^* E_p / 2\hbar$ and $\Delta_p^* = \omega_p^* - \omega_x$. In addition, a damping rate γ has been introduced to describe the very short lifetime of the virtual excitations.

Since the test field is in most cases much weaker than the pump field, we can safely apply perturbation theory to solve (5). The zeroth-order equations are obtained by setting $E_t = 0$ in (5), namely,

$$\dot{C}_+^0 = i\Omega_p^* e^{-i\Delta_p t} C_-^0 - \gamma C_+^0 \quad (6a)$$

$$\dot{C}_-^0 = i\Omega_p^* e^{i\Delta_p t} C_+^0 . \quad (6b)$$

In what follows, we assume for simplicity $\mu_p^* = \mu^*$ and $\Omega_p^* = \Omega_p^* t$. With the initial conditions $C_+^0(0) = 0$ and $C_-^0(0) = 1$, we find the solutions

$$C_+^0 = \frac{i\Omega_p}{\Omega} \sin\Omega_p t \exp[-(i\Delta_p + \gamma)t/2] \quad (7a)$$

$$C_-^0 = (\cos\Omega_p t + \frac{\gamma - i\Delta_p}{2\Omega} \sin\Omega_p t) e^{(i\Delta_p - \gamma)t/2} , \quad (7b)$$

where we have defined the complex Rabi frequency

$$\Omega = \frac{1}{2} [(\Delta_p + i\gamma)^2 + 4\Omega_p^2]^{1/2} . \quad (8)$$

The real part of the Rabi frequency is given by

$$\Omega_R = [(\sqrt{x^2 + y^2} + x)/8]^{1/2} , \quad (9a)$$

which determines the oscillation frequency, and the imaginary part by

$$\Omega_i = -[(\sqrt{x^2 + y^2} - x)/8]^{1/2} , \quad (9b)$$

which determines the envelope of the oscillation. In (9) we have defined $x = \Delta_p^2 + 4\Omega_p^2 - \gamma^2$ and $y = 2\Delta_p \gamma$, with $\Delta_p < 0$.

The solutions up to first-order perturbation can be obtained by inserting the zeroth-order solutions (7) into the right-hand side of (5). The results are

$$C_{\pm}(t) = C_{\pm}^0(t) + \delta C_{\pm}(t) \pm A_{\pm} \quad (10)$$

$$\begin{aligned} \delta C_{-} &= \frac{\Omega_t \Omega_p}{\Omega} \frac{\exp[i\Delta_t t - \frac{1}{2}(i\Delta_p + \gamma)t]}{[\frac{1}{2}(i\Delta_p + \gamma) - i\Delta_t]^2 + \Omega^2} \\ &\times [(\frac{1}{2}(i\Delta_p + \gamma) - i\Delta_t) \sin\Omega t + \Omega \cos\Omega t] \end{aligned} \quad (11a)$$

$$\begin{aligned} \delta C_{+} &= \frac{i\Omega_t}{2\Omega} \frac{\exp[-i\Delta_t t + \frac{1}{2}(i\Delta_p - \gamma)t]}{[i\Delta_t - \frac{1}{2}(i\Delta_p - \gamma)]^2 + \Omega^2} \\ &\times [(\Delta_p^2 + i\gamma)^2 + 2\Omega_p^2 + i\Delta_t(i\Delta_p - \gamma)] \sin\Omega t \\ &- 2\Omega[i(\Delta_t - \Delta_p) + \gamma] \cos\Omega t \end{aligned} \quad (11b)$$

The integration constants A_{\pm} in (10) are determined by the initial conditions.

The expectation value of the dipole moment of a virtual exciton is

$$\begin{aligned} \mu(t) &= \mu_0 \langle S^-(t) \rangle e^{-i\omega_x t} = \mu_0 C_{-}^* C_{+} e^{i\omega_x t} \\ &= \mu_0 (C_{-}^{0*} + \delta C_{-}^* + A_{-}^*)(C_{+}^0 + \delta C_{+} + A_{+}) e^{-i\omega_x t} \end{aligned} \quad (12)$$

It is evident that the dipole moment induced by the test beam is given by

$$\delta\mu(t) = \mu_0 (C_{-}^{0*} \delta C_{+} + C_{+}^0 \delta C_{-}^*) e^{-i\omega_x t} \quad (13)$$

Therefore, the nonlinear optical susceptibility experienced by the test beam is simply

$$\begin{aligned} x_t &= n\delta\mu(t)/(E_t e^{i\omega_t t}) \\ &= in \frac{|\mu_0|^2}{2\hbar} e^{-\gamma t} \left(\frac{1}{2\Omega z_1} [\cos\Omega^* t + i \frac{\Delta_p - i\gamma}{2\Omega^*} \sin\Omega^* t] \right. \\ &\times [(\Delta_p + i\gamma)^2 + 2\Omega_p^2 - \Delta_t(\Delta_p + i\gamma)] \sin\Omega t \\ &- 2\Omega[i(\Delta_t - \Delta_p) + \gamma] \cos\Omega t \\ &\left. - \frac{\Omega^2}{|\Omega|^2 z_2} \sin\Omega t [(\frac{1}{2}(i\Delta_p - \gamma) - i\Delta_t) \sin\Omega^* t + \Omega^* \cos\Omega^* t] \right) \end{aligned} \quad (14)$$

where

$$z_1 = \left[\frac{1}{2} (i\Delta_p - \gamma) - i\Delta_t \right]^2 + \Omega^2 \quad (15a)$$

$$z_2 = \left[\frac{1}{2} (i\Delta_p - \gamma) - i\Delta_t \right]^2 + \Omega^*{}^2 \quad (15b)$$

We have introduced in (14) the optically-induced transient virtual exciton density n which may be assumed to obey the rate equation²³

$$\frac{dn}{dt} = \eta\alpha I_p / (\hbar\omega_p) - \gamma n \quad (16)$$

where η denotes the quantum efficiency of virtual exciton generation per absorbed photon, $I_p = E_p c n_0 / 8\pi$ is the pumping intensity with n_0 as the linear refractive index, and α represents the absorption coefficient which is assumed to be a constant in the present case. In practice, however, α may be related to the density n to some extent.

Equation (16) can be solved by a solution of the form

$$n = n_s (1 - e^{-\gamma t}) \quad (17)$$

where $n_s = \eta\alpha I_p / (\hbar\omega_p)$. Once again, the initial condition $n(0) = 0$ is already assumed. The transient behavior of the nonlinear optical susceptibility x_t is calculated numerically as a function of time and the detuning Δ_t . The results are presented and discussed in the next section.

2.2 Optical nutation and hole burning

In our numerical study, we consider PTS and use the unit $\hbar = 1$. Other parameters are chosen as¹²⁻¹⁴ $\Delta_p = 0.1$ eV, $\Omega_p = 0.02$ eV, $\gamma = 0.01$ eV, $\frac{1}{2}|\mu|^2 n_s = \frac{1}{40\pi}$ eV and $\omega_p = 2$ eV. The real and imaginary parts of x_t are computed separately. Figure 1 shows snapshots at different instants of time of the variation of x_t with the detuning Δ_t in the unit of eV. The variation with time for fixed detuning is shown in Fig. 2.

It is clearly seen from Fig. 1 that both the differential absorption and refraction are direct effects of the virtual excitons induced by the pumping field. Nonzero susceptibility starts to show up as $|\Delta_t| \rightarrow 0.1$ and approaches its maximum as $\Delta_t \rightarrow 0$, where the test field is at resonance with the exciton. This indicates that there is energy transfer from the pump field to the test field via virtual excitons, in qualitative agreement with experiments.^{12,13}

Optical nutation can be observed in polymers in the ultrashort time regime when virtual excitons are generated by a pump field tuned below the exciton resonance. This situation can be seen from Fig. 2. It is also clearly shown that the oscillation centers of both $\text{Re}x_t$ and $\text{Im}x_t$ change with time. This is quite different from nutation phenomena predicted in Refs. 21 and 23 for semiconductors, where the oscillation amplitude decreases monotonically like damped oscillators.

Since it takes time for the pump field to build up enough virtual excitons and since the virtual excitons are short-lived, their effects are appreciable and stable most of the time during the pulse (on the order of picoseconds). The response time can be anywhere between 10 ~ 100 femtoseconds according to our calculation. Both

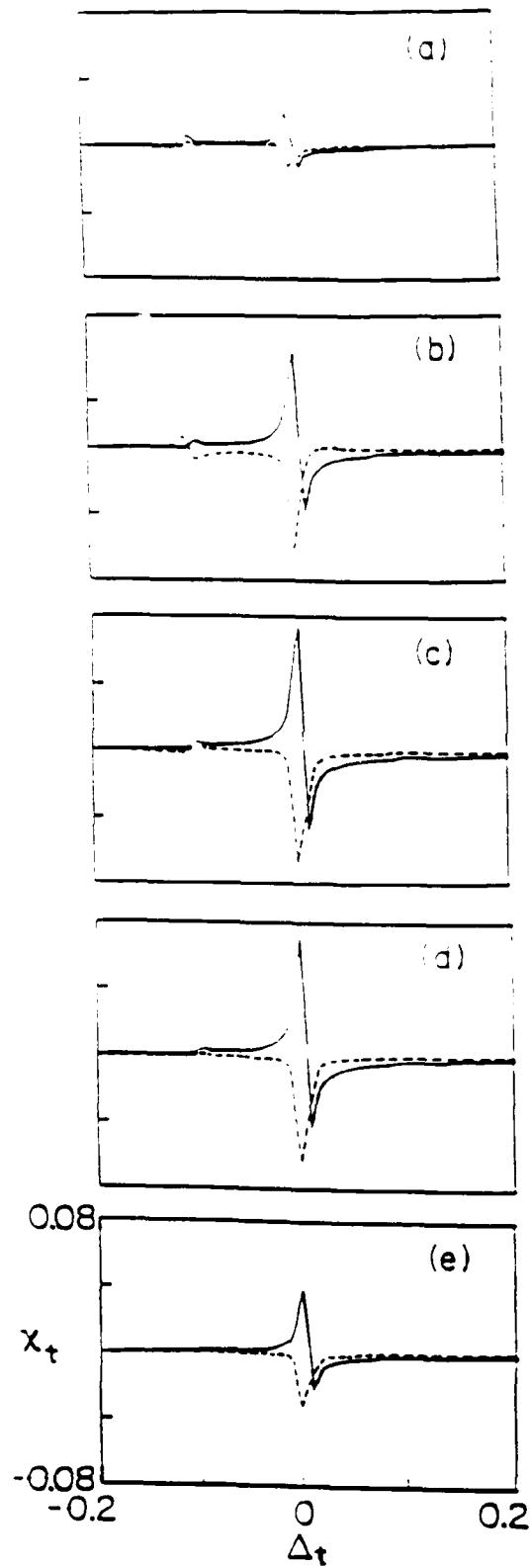


Figure 1. Snapshots of the real (solid lines) and imaginary (dashed line) parts of the nonlinear susceptibility at various times: (a) $t = 25 \text{ eV}^{-1}$, (b) $t = 100 \text{ eV}^{-1}$, (c) $t = 250 \text{ eV}^{-1}$, (d) $t = 500 \text{ eV}^{-1}$ and (e) $t = 1500 \text{ eV}^{-1}$.

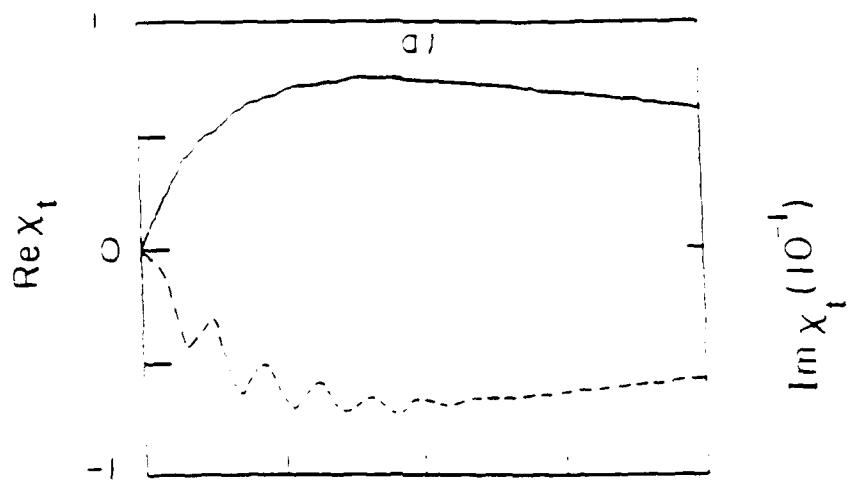
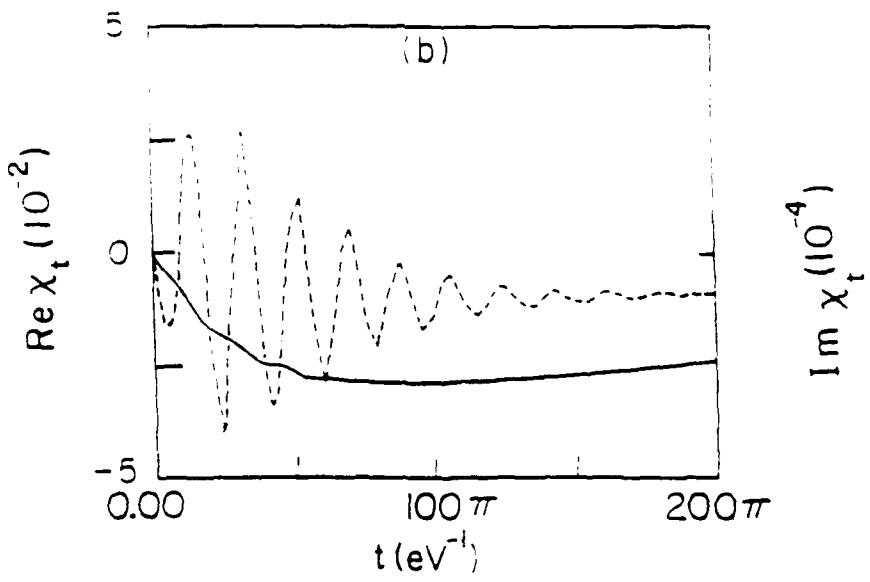


Figure 2. Real (solid lines) and imaginary (dashed lines) parts of χ_t versus time: (a) $\Delta_t = 0$ and (b) $\Delta_t = 0.1$ eV.



the absorption and refraction parts show up almost right away and taper off rather slowly after reaching their peaks.

In addition, we can also observe from these results that increasing the intensity of the pump field leads to a deeper and wider hole in the absorption spectrum, which is qualitatively in agreement with the steady-state results of Ref. 10. What seems to be more interesting is that we find a blue shift of 0.005 eV in our calculation, even though we have not considered the effect of phonons. A more careful treatment with phonon mediation included can be found in Ref. 22. This is the electronic state optical Stark shift observed in a recent experiment¹⁴ in which the differential transmission of polydiacetylene-toluene sulfonate has been measured by means of coherent inverse Raman spectroscopy. It is demonstrated in Ref. 14 that this ac Stark effect is important in the determination of the nonlinear optical response of PTS even in the small signal limit. The effect is reproduced theoretically in Ref. 14 by including an extra term to modify the exciton resonance frequency in the simple model of Schmitt-Rink et al.¹³ However, the treatment in both Refs. 13 and 14 assumes a steady-state solution, while the experimental conditions are more transient-like.

3. PHONON-MEDIATED OPTICAL TRISTABILITY IN PTS

We now turn our attention to a special case in which a sample of PTS is placed inside an optical cavity, and investigate its response to a laser beam directed into the cavity. In particular, we look for the dependence of the output field intensity on the intensity of the incident laser field. To describe our system, we assume the model Hamiltonian of Ref. 13 with an extra term representing the cavity photon-exciton interaction. Thus

$$H = \Omega a^\dagger a + \omega_{ph} b^\dagger b + \omega_{ex} c^\dagger c + \lambda c^\dagger c(b + b^\dagger) + ig(a^\dagger c - c^\dagger a) + i(a^\dagger E e^{-i\Omega t} - a E^* e^{i\Omega t}), \quad (18)$$

where a^\dagger (a), b^\dagger (b) and c^\dagger (c) are the creation (annihilation) operators for the cavity field, phonon and exciton, respectively, with corresponding frequencies Ω , ω_{ph} and ω_{ex} . The phonon-exciton coupling constant is denoted by λ , and the exciton-field coupling constant by g . The coherent driving field is assumed to have the same frequency Ω , and its amplitude is E whose spatial dependence is ignored completely. For simplicity, we have included only one of the phonon modes²⁴ which couples most strongly to the exciton, with momentum dependence neglected.¹³

In the usual zero-fluctuation limit, quantum mechanical operators can be replaced by their expectation values $\alpha = \langle a \rangle$, $\beta = \langle b \rangle$, $\eta = \langle c \rangle$ and $n_c = \langle c^\dagger c \rangle$, and in the limit of high Q cavity, the phonon and exciton variables are eliminated adiabatically.²⁵ Without going into the details which will be published elsewhere,²⁶ we discuss only briefly the results here. The equation of motion for the field variable, expressed in the unit such that $\gamma_{ex} = 1$, takes the form

$$\frac{\partial}{\partial \tau} \alpha_c = \sqrt{2g^2} E - \alpha_c f(|\alpha_c|^2), \quad (19)$$

where the function f is defined as

$$f(x) = 1 + \left(\frac{g^2}{\kappa}\right) \frac{1 - i[\Delta - \lambda_p n_c(x)]}{1 + [\Delta - \lambda_p n_c(x)]^2} \quad (20)$$

In Eq. (19), we have introduced for convenience the phenomenological damping rates κ , γ and γ_{ph} and dimensionless variables $\tau = \kappa t$, $E = E/\kappa$ and $\alpha = \frac{\gamma}{\Delta} g \alpha_0$. The field intensity is expressed as $I = |\alpha_c|^2$ and the detuning as $\Delta = \omega_{\text{ex}} - \Omega$.

In the steady state, we set $\alpha_c = \alpha_s = \text{constant}$ in (19) and (20). The steady-state cavity field intensity can then be found straightforwardly as

$$I_s = -\frac{1 + Y_s^2(I_{\text{in}})}{(1 + g^2/\kappa)^2 + Y_s^2(I_{\text{in}})} I_{\text{in}} \quad (21)$$

where we have defined the driving field intensity $I_{\text{in}} = 2g^2|E|^2$ and $Y_s(I_{\text{in}}) = \Delta - \lambda_p n_s(I_{\text{in}})$ with $\lambda_p = 2\lambda^2/\omega_{\text{ph}}$. It can be shown from (21) that the steady-state virtual exciton n_s satisfies the equation

$$n_s^3 - 2(\Delta/\lambda_p)^2 n_s^2 + [\Delta^2 + (1 + g^2/\kappa)^2] \lambda_p^{-2} n_s - I_{\text{in}} \lambda_p^{-2} = 0 \quad (22)$$

When the conditions are such that Eq. (22) has three distinct roots, there will be three output I_s for each given I_{in} . As has been discussed in Ref. 26, the conditions of multi-real roots of (22) require that tristability occurs only when the cavity field is tuned sufficiently below the exciton resonance and when there exists phonon-exciton coupling. This clearly shows that the tristability is mediated by phonons.

Numerical calculation of Eq. (21) has been performed for PTS for which^{13,14} $\omega_{\text{ph}} = 5.16$ and $\lambda = 2.0$. The results are plotted in Fig. 3 for various choices of Δ and g^2/κ . It is found that all the multiple solutions are stable except for those around the turning points A and B in Figs. 3(b) and 3(c). As I_{in} increases continuously, passes the turning point and then decreases, the output I_s will follow a reversed hysteresis loop. The middle branch can only be reached if I_{in} reverses its course of change at the turning point.

It is emphasized that the dispersion effect plays a crucial role in this kind of tristability. When I_{in} increases beyond a certain level, n_s becomes large enough that the lattice relaxation is sufficiently high for the refractive index to deviate from the cavity resonance. Consequently, the cavity field switches to the lower branch. On the other hand, such tristability cannot occur when the Q-value of the optical cavity is excessively high. As a final remark, we remark that this tristability is found by considering only one phonon mode in PTS. It is therefore expected to find optical multistability if more phonon modes are included.

ACKNOWLEDGMENTS

This research was supported by the Office of Naval Research.

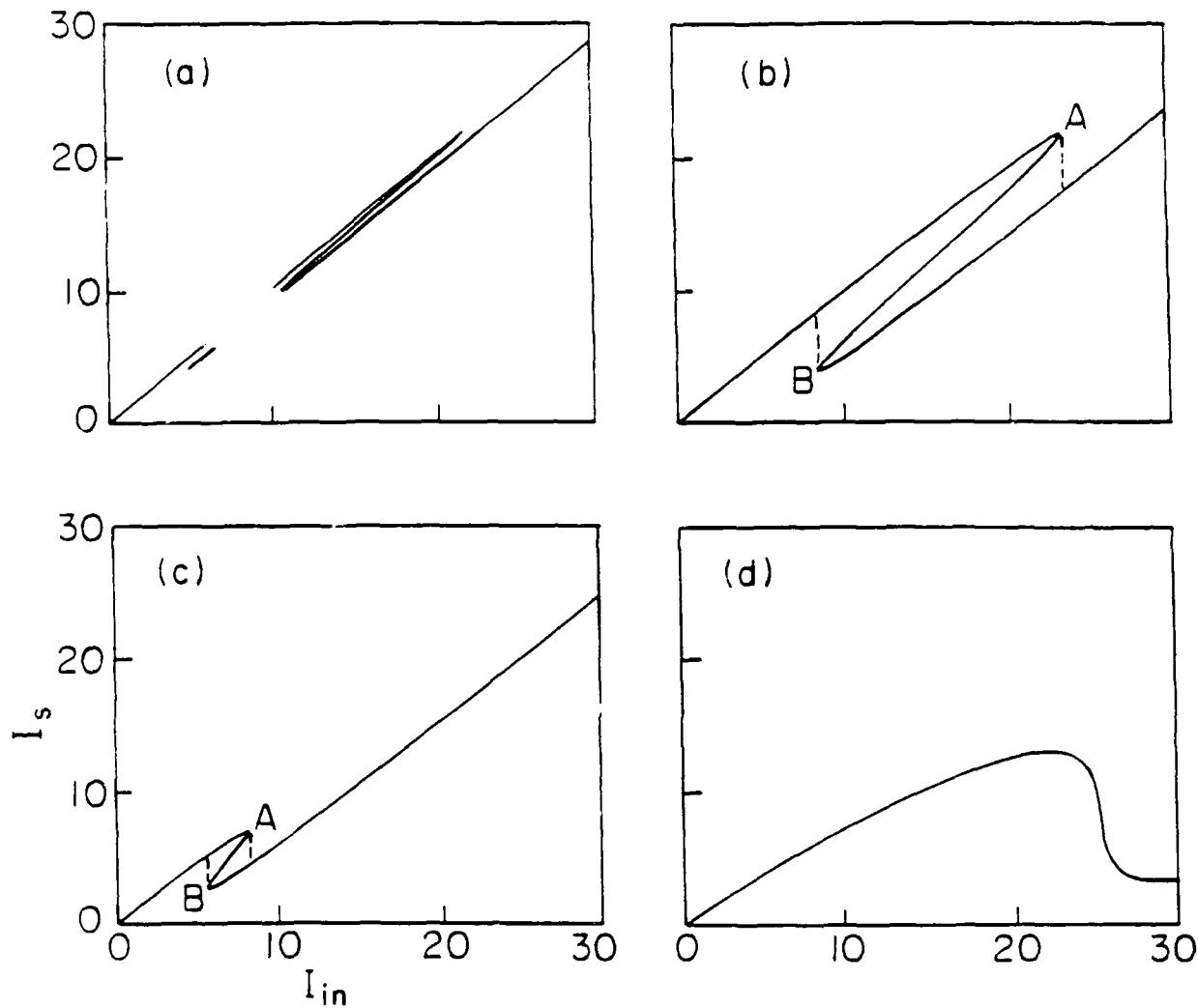


Figure 3. Intensity of the cavity field I_s versus the input field intensity I_{in} : (a) $\Delta = 6$, $g^2/\kappa = 0.1$; (b) $\Delta = 6$, $g^2/\kappa = 0.5$; (c) $\Delta = 4$, $g^2/\kappa = 0.5$; (d) $\Delta = 5$, $g^2/\kappa = 2$. The dashed lines in (b) and (c) are hypothetical and drawn only to complete the reversed hysteresis loop.

REFERENCES

1. See, for example, D. J. Williams, in Electronic and Photonic Applications of Polymers, ed. by M. J. Bowden and S. R. Turner (American Chemical Society, Washington, D.C.), ACS Advances in Chem. 218, 297 (1988).
2. D. S. Chemla and J. Zyss, Eds., Nonlinear Optical Properties of Organic Molecules and Crystals (Academic Press, Orlando, 1987).
3. J. P. Hermann, D. Richard and J. Ducuing, Appl. Phys. Lett. 23, 178 (1973); K. C. Rustagi and J. Ducuing, Opt. Commun. 10, 258 (1974); C. Sauteret, J. P. Hermann, R. Frey, F. Pradere, J. Ducuing, R. H. Baughman and R. R. Chance, Phys. Rev. Lett. 36, 956 (1976).
4. F. Kajzar, S. Etemad, G. L. Baker and J. Messier, Synth. Met. 17, 563 (1987); Solid State Commun. 63, 1113 (1987); F. Krausz, E. Wintner and G. Leising, Phys. Rev. B 39, 3701 (1989).
5. W. P. Su, J. R. Schrieffer and A. J. Heeger, Phys. Rev. B 22, 2099 (1980).
6. H. Takayama, Y. R. Lin-Liu and K. Maki, Phys. Rev. B 21, 2388 (1986).
7. W. Wu, Phys. Rev. Lett. 61, 1119 (1988).
8. D. M. Mackie, R. J. Cohen and A. J. Glick, Phys. Rev. B 38, 3442 (1989).
9. J. Yu, B. Friedman, P. R. Baldwin and W. P. Su, Phys. Rev. B 39, 12814 (1989).
10. G. M. Carter, Y. J. Chen and S. Tripathy, Appl. Phys. Lett. 43, 892 (1983).
11. M. Sinclair, D. McBranch, D. Moses and A. J. Heeger, Appl. Phys. Lett. 53, 2374 (1988).
12. B. I. Greene, J. Orenstein, R. R. Millard and L. R. Williams, Chem. Phys. Lett. 139, 381 (1987); B. I. Greene, J. Orenstein, R. R. Millard and L. R. Williams, Phys. Rev. Lett. 58, 2750 (1987); B. I. Green, J. Orenstein, M. Tahkur and D. H. Rapkine, Mat. Res. Soc. Symp. Proc. 109, 159 (1987).
13. B. I. Greene, J. F. Mueller, J. Orenstein, D. H. Rapkine, S. Schmitt-Rink and M. Thakur, Phys. Rev. Lett. 61, 325 (1988).
14. G. J. Blanchard, J. P. Heritage, A. C. Von Lehmen, G. L. Baker and S. Etemad, Bull. Am. Phys. Soc. 34, 452 (1989); Phys. Rev. Lett. 63, 887 (1989).
15. A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migns, W. T. Masselink and H. Morkoc, Phys. Rev. Lett. 56, 2748 (1986).
16. S. Schmitt-Rink and D. S. Chemla, Phys. Rev. Lett. 57, 2752 (1986).

17. A. Von Lenman, D. S. Chemla, J. E. Zucker and J. P. Heritage, Opt. Lett. 11, 609 (1986).
18. D. S. Chemla, D. A. B. Miller and S. Schmitt-Rink, in Optical Nonlinearities and Instabilities in Semiconductors (Academic, New York, 1988).
19. P. D. Townsend, G. L. Baker, N. E. Schlottter and S. Etemad, Synth. Met. 28, D633 (1989).
20. M. Thakur, Y. Shani, C. C. Chi and K. O'Brian, Synth. Met. 28, D595 (1989).
21. P. Sen and P. K. Sen, IEEE J. Quantum Electron. QE23, 2033 (1987).
22. X. S. Li, D. L. Lin, T. F. George and X. Sun, Phys. Rev. B 40, in press.
23. K. Singh, P. Sen and P. K. Sen, IEEE J. Quantum Electron. QE25, 67 (1989).
24. See D. N. Batchelder, in Polydiacetylenes, ed. by D. Bloor and R. R. Chance (Martinus Nijhoff, Dordrecht, the Netherlands, 1985), p. 187 ff.
25. M. Reid, K. J. McNeil and D. F. Walls, Phys. Rev. A 24, 2029 (1981).
26. X. S. Li, D. L. Lin and T. F. George, Phys. Rev. B (Rapid Commun.), in press.

AL/1113/86/2

TECHNICAL REPORT DISTRIBUTION LIST, GEN

	<u>No.</u> <u>Copies</u>		<u>No.</u> <u>Copies</u>
Office of Naval Research Attn: Code 1113 800 N. Quincy Street Arlington, Virginia 22217-5000	2	Dr. David Young Code 334 NORDA NSTL, Mississippi 39529	1
Dr. Bernard Douda Naval Weapons Support Center Code 50C Crane, Indiana 47522-5050	1	Naval Weapons Center Attn: Dr. Ron Atkins Chemistry Division China Lake, California 93555	1
Naval Civil Engineering Laboratory Attn: Dr. R. W. Drisko, Code L52 Port Hueneme, California 93401	1	Scientific Advisor Commandant of the Marine Corps Code RD-1 Washington, D.C. 20380	1
Defense Technical Information Center Building 5, Cameron Station Alexandria, Virginia 22314	12 high quality	U.S. Army Research Office Attn: CRD-AA-IP P.O. Box 12211 Research Triangle Park, NC 27709	1
DTNSRDC Attn: Dr. H. Singerman Applied Chemistry Division Annapolis, Maryland 21401	1	Mr. John Boyle Materials Branch Naval Ship Engineering Center Philadelphia, Pennsylvania 19112	1
Dr. William Tolles Superintendent Chemistry Division, Code 6100 Naval Research Laboratory Washington, D.C. 20375-5000	1	Naval Ocean Systems Center Attn: Dr. S. Yamamoto Marine Sciences Division San Diego, California 91232	1
		Dr. David L. Nelson Chemistry Division Office of Naval Research 800 North Quincy Street Arlington, Virginia 22217	1

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. J. E. Jensen
Hughes Research Laboratory
3011 Malibu Canyon Road
Malibu, California 90265

Dr. C. B. Harris
Department of Chemistry
University of California
Berkeley, California 94720

Dr. J. H. Weaver
Department of Chemical Engineering
and Materials Science
University of Minnesota
Minneapolis, Minnesota 55455

Dr. F. Kutzler
Department of Chemistry
Box 5055
Tennessee Technological University
Cookeville, Tennessee 38501

Dr. A. Reisman
Microelectronics Center of North Carolina
Research Triangle Park, North Carolina
27709

Dr. D. Dilella
Chemistry Department
George Washington University
Washington D.C. 20052

Dr. M. Grunze
Laboratory for Surface Science and
Technology
University of Maine
Orono, Maine 04469

Dr. R. Reeves
Chemistry Department
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. J. Butler
Naval Research Laboratory
Code 6115
Washington D.C. 20375-5000

Dr. Steven M. George
Stanford University
Department of Chemistry
Stanford, CA 94305

Dr. L. Interante
Chemistry Department
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. Mark Johnson
Yale University
Department of Chemistry
New Haven, CT 06511-8118

Dr. Irvin Heard
Chemistry and Physics Department
Lincoln University
Lincoln University, Pennsylvania 19352

Dr. W. Knauer
Hughes Research Laboratory
3011 Malibu Canyon Road
Malibu, California 90265

Dr. K.J. Klaubunde
Department of Chemistry
Kansas State University
Manhattan, Kansas 66506

2/1113/86/2

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. G. A. Somorjai
Department of Chemistry
University of California
Berkeley, California 94720

Dr. J. Murday
Naval Research Laboratory
Code 6170
Washington, D.C. 20375-5000

Dr. J. B. Hudson
Materials Division
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. Theodore E. Madey
Surface Chemistry Section
Department of Commerce
National Bureau of Standards
Washington, D.C. 20234

Dr. J. E. Demuth
IBM Corporation
Thomas J. Watson Research Center
P.O. Box 218
Yorktown Heights, New York 10598

Dr. M. G. Lagally
Department of Metallurgical
and Mining Engineering
University of Wisconsin
Madison, Wisconsin 53706

Dr. R. P. Van Duyne
Chemistry Department
Northwestern University
Evanston, Illinois 60637

Dr. J. M. White
Department of Chemistry
University of Texas
Austin, Texas 78712

Dr. D. E. Harrison
Department of Physics
Naval Postgraduate School
Monterey, California 93940

Dr. R. L. Park
Director, Center of Materials
Research
University of Maryland
College Park, Maryland 20742

Dr. W. T. Peria
Electrical Engineering Department
University of Minnesota
Minneapolis, Minnesota 55455

Dr. Keith H. Johnson
Department of Metallurgy and
Materials Science
Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

Dr. S. Sibener
Department of Chemistry
James Franck Institute
5640 Ellis Avenue
Chicago, Illinois 60637

Dr. Arnold Green
Quantum Surface Dynamics Branch
Code 3817
Naval Weapons Center
China Lake, California 93555

Dr. A. Wold
Department of Chemistry
Brown University
Providence, Rhode Island 02912

Dr. S. L. Bernasek
Department of Chemistry
Princeton University
Princeton, New Jersey 08544

Dr. W. Kohn
Department of Physics
University of California, San Diego
La Jolla, California 92037

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. F. Carter
Code 61/0
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. Richard Colton
Code 6170
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. Dan Pierce
National Bureau of Standards
Optical Physics Division
Washington, D.C. 20234

Dr. R. Stanley Williams
Department of Chemistry
University of California
Los Angeles, California 90024

Dr. R. P. Messmer
Materials Characterization Lab.
General Electric Company
Schenectady, New York 22217

Dr. Robert Gomer
Department of Chemistry
James Franck Institute
5640 Ellis Avenue
Chicago, Illinois 60637

Dr. Ronald Lee
R301
Naval Surface Weapons Center
White Oak
Silver Spring, Maryland 20910

Dr. Paul Schoen
Code 6190
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. John T. Yates
Department of Chemistry
University of Pittsburgh
Pittsburgh, Pennsylvania 15260

Dr. Richard Greene
Code 5230
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. L. Kesmodel
Department of Physics
Indiana University
Bloomington, Indiana 47403

Dr. K. C. Janda
University of Pittsburgh
Chemistry Building
Pittsburg, PA 15260

Dr. E. A. Irene
Department of Chemistry
University of North Carolina
Chapel Hill, North Carolina 27514

Dr. Adam Heller
Bell Laboratories
Murray Hill, New Jersey 07974

Dr. Martin Fleischmann
Department of Chemistry
University of Southampton
Southampton 509 5NH
UNITED KINGDOM

Dr. H. Tachikawa
Chemistry Department
Jackson State University
Jackson, Mississippi 39217

Dr. John W. Wilkins
Cornell University
Laboratory of Atomic and
Solid State Physics
Ithaca, New York 14853

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. R. G. Wallis
Department of Physics
University of California
Irvine, California 92664

Dr. D. Ramaker
Chemistry Department
George Washington University
Washington, D.C. 20052

Dr. J. C. Hemminger
Chemistry Department
University of California
Irvine, California 92717

Dr. T. F. George
Chemistry Department
University of Rochester
Rochester, New York 14627

Dr. G. Rubloff
IBM
Thomas J. Watson Research Center
P.O. Box 218
Yorktown Heights, New York 10598

Dr. Horia Metiu
Chemistry Department
University of California
Santa Barbara, California 93106

Dr. W. Goddard
Department of Chemistry and Chemical
Engineering
California Institute of Technology
Pasadena, California 91125

Dr. P. Hansma
Department of Physics
University of California
Santa Barbara, California 93106

Dr. J. Baldeschwieler
Department of Chemistry and
Chemical Engineering
California Institute of Technology
Pasadena, California 91125

Dr. J. T. Keiser
Department of Chemistry
University of Richmond
Richmond, Virginia 23173

Dr. R. W. Plummer
Department of Physics
University of Pennsylvania
Philadelphia, Pennsylvania 19104

Dr. E. Yeager
Department of Chemistry
Case Western Reserve University
Cleveland, Ohio 41106

Dr. N. Winograd
Department of Chemistry
Pennsylvania State University
University Park, Pennsylvania 16802

Dr. Roald Hoffmann
Department of Chemistry
Cornell University
Ithaca, New York 14853

Dr. A. Steckl
Department of Electrical and
Systems Engineering
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. G.H. Morrison
Department of Chemistry
Cornell University
Ithaca, New York 14853